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(54) [Title of the Invention]

INDICATOR FOR INSECTICIDES

(57) [Abstract]

[Constitution] An indicator for insecticides which is characterized by the fact that said indicator contains a volatilizable desensitizing agent, an electron-donative coloring agent and an inorganic developing compound as essential components, and the aforementioned volatilizable desensitizing agent is preferably a pyrethroid that is volatilized at ordinary temperatures.

[Effect] The indicator for insecticides provided by the present invention makes it possible to detect very clearly the volatilization process and endpoint of the volatilization process of the [abovementioned] pyrethroid volatilizable at ordinary temperatures, which constitutes the [abovementioned] volatilizable desensitizing agent. Furthermore, this indicator combines a high insecticidal/insect-proofing effect, a low toxicity for warm-blooded animals, and [good] chemical stability. Accordingly, the practical utility of this indicator is extremely high.

[Claims]

[Claim 1] An indicator for insecticides which is characterized by the fact that said indicator contains a volatilizable desensitizing agent, an electron-donative coloring agent and an inorganic developing compound as essential components.

[Claim 2] The indicator for insecticides claimed in Claim 1, which is characterized by the fact that the [aforementioned] volatilizable desensitizing agent and electron-donative coloring agent are applied by impregnation, injection or coating to a holding body consisting of or containing an inorganic developing compound, or are held by such a holding body.

[Claim 3] The indicator for insecticides claimed in Claim 1 or Claim 2, which is characterized by the fact that the aforementioned volatilizable desensitizing agent is a pyrethroid which is volatilizable at ordinary temperatures.

[Claim 4] The indicator for insecticides claimed in Claim 3, which is characterized by the fact that the aforementioned pyrethroid that is volatilizable at ordinary temperatures consists of one or two compounds selected from a set consisting of empentrin [translit.—Tr.], 5-propargyl-2-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate, 5-propargyl-2-methyl-3-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate and benfluthrin [?] [translit.—Tr.].

[Claim 5] The indicator for insecticides claimed in Claim 3 or Claim 4, which is characterized by the fact that said indicator shows color development when the weight ratio of the volatilizable desensitizing agent to the inorganic developing compound is in the range of 30 : 1 ~ 2 : 1.

[Claim 6] The indicator for insecticides claimed in any of Claims 1 through 3, which is characterized by the fact that the inorganic developing compound consists of one or more compounds selected from a set consisting of silica, activated alumina, clay, talc, powdered quartzite (powdered silica) [?] [doubtful wording—Tr.], acid clay, activated clay, bentonite, kaolin, cerite [?] [translit.—Tr.], fluorigel [?] [translit.—Tr.], pearlite, aluminum silicate, magnesium silicate, titanium oxide and zinc oxide.

[Claim 7] The indicator for insecticides claimed in Claim 6, which is characterized by the fact that the inorganic developing compound consists of one or more compounds selected from a set consisting of silica, activated alumina and fluorigel [?].

[Claim 8] The indicator for insecticides claimed in Claim 5, which is characterized by the fact that said indicator contains a pyrethroid that is volatilizable at ordinary temperatures as the abovementioned volatilizable desensitizing agent, and one or more compounds selected from a set consisting of silica, activated alumina, clay, talc, powdered quartzite (powdered silica) [?] [doubtful wording—Tr.], acid clay, activated clay, bentonite, kaolin, cerite [?] [translit.—Tr.], fluorigel [?] [translit.—Tr.], pearlite, aluminum silicate, magnesium silicate, titanium oxide and zinc oxide as the abovementioned inorganic developing compound.

[Claim 9] The indicator for insecticides claimed in Claim 8, which is characterized by the fact that the aforementioned pyrethroid that is volatilizable at ordinary temperatures consists of one

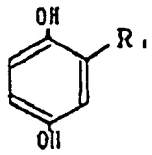
or two compounds selected from a set consisting of empentrin [translit.—Tr.], 5-propargyl-2-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate, 5-propargyl-2-methyl-3-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate and benfluthrin [?] [translit.—Tr.].

[Claim 10] The indicator for insecticides claimed in Claim 8 or Claim 9, which is characterized by the fact that the inorganic developing compound consists of one or more compounds selected from a set consisting of silica, activated alumina and fluorigel [?].

[Claim 11] The indicator for insecticides for insecticides claimed in Claim 9 or Claim 10, which is characterized by the fact that [a] the abovementioned pyrethroid that is volatilizable at ordinary temperatures consists of one or two compounds selected from a set consisting of empentrin [translit.—Tr.], 5-propargyl-2-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate, 5-propargyl-2-methyl-3-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate and benfluthrin [?] [translit.—Tr.], [b] the abovementioned inorganic developing compound consists of one or more compounds selected from a set consisting of silica, activated alumina and fluorigel [?], [c] the pyrethroid and developing compounds are mixed at a mixture ratio in the range of 100 : 1 ~ 5 : 1, and [the indicator] shows developing properties together with the volatilization of the pyrethroid that is volatilizable at ordinary temperatures in the [mixture ratio] range of 30 : 1 ~ 2 : 1.

[Claim 12] The indicator for insecticides claimed in any of Claims 8 through 11, which is characterized by the fact that said indicator contains a pyrethroid that is volatilizable at ordinary temperatures as the abovementioned volatilizable desensitizing agent, and said indicator also contains one or more hydroquinone type compounds expressed by general formula I

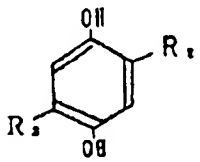
[Chemical Formula 1]



(in the above formula, R_1 indicates an alkyl group with 3 to 6 carbon atoms) as a stabilizer at the rate of 0.01 to 0.1 times the amount of the aforementioned pyrethroid that is volatilizable at ordinary temperatures.

[Claim 13] An indicator for insecticides which is characterized by the fact that in the indicator for insecticides claimed in Claim 12, one or more hydroquinone type compounds expressed by general formula II

[Chemical Formula 2]



(in the above formula, R_2 and R_3 indicate alkyl groups with 3 to 6 carbon atoms, which may be the same or different) are additionally added as stabilizers at the rate of 0.005 to 0.05 times the amount of the aforementioned pyrethroid that is volatilizable at ordinary temperatures.

[Claim 14] The indicator for insecticides claimed in any of Claims 8 through 13, which is characterized by the fact that a volatilization-adjusting agent for the pyrethroid that is volatilizable at ordinary temperatures is mixed [with the indicator].

[Claim 15] The indicator for insecticides claimed in Claim 1 or Claim 2, which is characterized by the fact that said indicator contains a colored die and/or a colored pigment along with the aforementioned electron-donative coloring agent as a coloring substance.

[Detailed Description of the Invention]

[0001]

[Field of Industrial Utilization] The present invention relates to an indicator for insecticides which is devised so that the volatilization process and endpoint of the volatilization process of a volatilizable agent can be confirmed visually and in an extremely clear manner by means of a color change.

[0002]

[Prior Art] Conventionally, various methods have been proposed for detecting the volatilization process of components accommodated in an accommodating body; the following methods may be cited as examples:

(1) Methods which use a vaporizable (volatilizable) dye, and which utilize the disappearance of color caused by the volatilization of this dye.

(2) Methods which use an indicator that causes a color change as a result of contact with CO_2 or H_2O in the atmosphere.

(3) Methods which combine an electron-donative coloring agent, an electron-accepting organic developing agent and a volatilizable desensitizing component, and which utilize coloring caused by the volatilization of the desensitizing component.

However, in the case of methods of type (1), the volatilization process and the endpoint of this process tend to be unclear, so that the materials continue to be used even after the active component has disappeared, or so that the materials are discarded in spite of the fact that the active component remains in a considerable quantity so that the materials are still usable. In the case of methods of type (2), the color change is influenced by environmental conditions such as the temperature and humidity at the location of use, etc., so that there are problems in the correlation with the volatilization of the active components. Furthermore, methods of type (3) are similar to the method of the present invention; however, since a phenol, etc., is used as the organic developing agent, there may be cases (depending on the compound used) in which the

coloring agent again loses color after temporarily coloring as a result of degeneration or volatilization, etc. Furthermore, there may also be problems with the stability of the organic developing agent itself. Examples of methods that use an inorganic compound as a developing agent include the method described in Japanese Patent Application Kokai No. SHO 63-212364 (hereafter referred to as the "cited example"); however, there is no disclosure of an example in which a volatilizable desensitizing agent is used in an indicator for insecticides. Furthermore, according to an investigation conducted by the present inventors, the desensitizing agent/developing compound ratio at which coloring of the electron-donative coloring agent occurs varies greatly according to the types and combinations of inorganic developing compounds and volatilizable desensitizing agents used, so that there are optimal mixture proportions of inorganic developing compounds for respective volatilizable desensitizing agents. Nevertheless, there is no mention of these facts in the cited example.

[0003]

[Problems to Be Solved by the Invention] The object of the present invention is to provide an indicator for insecticides which allows the very clear visual detection of the volatilization process and endpoint of the volatilization process of a volatilizable agent, and which is highly safe.

[0004]

[Means Used to Solve the Abovementioned Problems] The invention of Claim 1 consists of an indicator for insecticides which is characterized by the fact that said indicator contains a volatilizable desensitizing agent, an electron-donative coloring agent and an inorganic developing compound as essential components.

[0005] The invention of Claim 2 is [characterized by the fact that] in the invention of Claim 1, the [aforementioned] volatilizable desensitizing agent and electron-donative coloring agent are applied by impregnation, injection or coating to a holding body consisting of or containing an inorganic developing compound, or are held by such a holding body.

[0006] The invention of Claim 3 is [characterized by the fact that] in the invention of Claim 1 or Claim 2, the aforementioned volatilizable desensitizing agent is a pyrethroid which is volatilizable at ordinary temperatures.

[0007] The invention of Claim 4 is [characterized by the fact that] in the invention of Claim 3, the aforementioned pyrethroid that is volatilizable at ordinary temperatures consists of one or two compounds selected from a set consisting of empentrin [translit.—Tr.], 5-propargyl-2-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate, 5-propargyl-2-methyl-3-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate and benfluthrin [?] [translit.—Tr.].

[0008] The invention of Claim 5 is [characterized by the fact that] in the invention claimed in Claim 3 or Claim 4, the indicator shows color development when the weight ratio of the volatilizable desensitizing agent to the inorganic developing compound is in the range of 30 : 1 ~ 2 : 1.

[0009] The invention of Claim 6 is [characterized by the fact that] in any of the inventions claimed in Claims 1 through 3, the inorganic developing compound consists of one or more compounds selected from a set consisting of silica, activated alumina, clay, talc, powdered quartzite (powdered silica) [?] [doubtful wording—Tr.], acid clay, activated clay, bentonite, kaolin, cerite [?] [translit.—Tr.], fluorigel [?] [translit.—Tr.], pearlite, aluminum silicate, magnesium silicate, titanium oxide and zinc oxide.

[0010] The invention of Claim 7 is [characterized by the fact that] in the invention claimed in Claim 6, the inorganic developing compound consists of one or more compounds selected from a set consisting of silica, activated alumina and fluorigel [?].

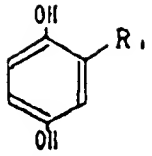
[0011] The invention of Claim 8 is [characterized by the fact that] in the invention claimed in Claim 5, the indicator contains a pyrethroid that is volatilizable at ordinary temperatures as the abovementioned volatilizable desensitizing agent, and one or more compounds selected from a set consisting of silica, activated alumina, clay, talc, powdered quartzite (powdered silica) [?] [doubtful wording—Tr.], acid clay, activated clay, bentonite, kaolin, cerite [?] [translit.—Tr.], fluorigel [?] [translit.—Tr.], pearlite, aluminum silicate, magnesium silicate, titanium oxide and zinc oxide as the abovementioned inorganic developing compound.

[0012] The invention of Claim 9 is [characterized by the fact that] in the invention claimed in Claim 8, the aforementioned pyrethroid that is volatilizable at ordinary temperatures consists of one or two compounds selected from a set consisting of empentrin [translit.—Tr.], 5-propargyl-2-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate, 5-propargyl-2-methyl-3-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate and benfluthrin [?] [translit.—Tr.].

[0013] The invention of Claim 10 is [characterized by the fact that] in the invention claimed in Claim 8 or Claim 9, the inorganic developing compound consists of one or more compounds selected from a set consisting of silica, activated alumina and fluorigel [?].

[0014] The invention of Claim 11 is [characterized by the fact that] in the invention claimed in Claim 9 or Claim 10, [a] the abovementioned pyrethroid that is volatilizable at ordinary temperatures consists of one or two compounds selected from a set consisting of empentrin [translit.—Tr.], 5-propargyl-2-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate, 5-propargyl-2-methyl-3-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate and benfluthrin [?] [translit.—Tr.], [b] the abovementioned inorganic developing compound consists of one or more compounds selected from a set consisting of silica, activated alumina and fluorigel [?], [c] the pyrethroid and developing compounds are mixed at a mixture ratio in the range of 100 : 1 ~ 5 : 1, and [d] [the indicator] shows developing properties together with the volatilization of the pyrethroid that is volatilizable at ordinary temperatures in the [mixture ratio] range of 30 : 1 ~ 2 : 1.

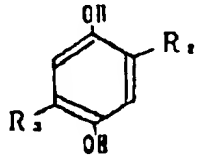
[0015] The invention of Claim 12 is [characterized by the fact that] in any of the inventions claimed in Claims 8 through 11, the indicator contains a pyrethroid that is volatilizable at ordinary temperatures as the abovementioned volatilizable desensitizing agent, and said indicator also contains one or more hydroquinone type compounds expressed by general formula I [Chemical Formula 3]



(in the above formula, R_1 indicates an alkyl group with 3 to 6 carbon atoms) as a stabilizer at the rate of 0.01 to 0.1 times the amount of the aforementioned pyrethroid that is volatilizable at ordinary temperatures.

[0016] The invention of Claim 13 is [characterized by the fact that] in the invention claimed in Claim 12, one or more hydroquinone type compounds expressed by general formula II

[Chemical Formula 4]



(in the above formula, R_2 and R_3 indicate alkyl groups with 3 to 6 carbon atoms, which may be the same or different) are additionally added as stabilizers at the rate of 0.005 to 0.05 times the amount of the aforementioned pyrethroid that is volatilizable at ordinary temperatures.

[0017] The invention of Claim 14 is [characterized by the fact that] in any of the inventions claimed in Claims 8 through 13, a volatilization-adjusting agent for the pyrethroid that is volatilizable at ordinary temperatures is mixed [with the indicator].

[0018] The invention of Claim 15 is [characterized by the fact that] in the invention claimed in Claim 1 or Claim 2, the indicator contains a colored die and/or a colored pigment along with the aforementioned electron-donative coloring agent as a coloring substance.

[0019]

[Operation] The indicator for insecticides of the invention claimed in Claim 1 is devised so that the volatilization process and endpoint of the volatilization process of the [aforementioned] volatilizable desensitizing agent can be visually detected very clearly and safely by the color change of an electron-donative coloring agent caused by an inorganic developing compound.

[0020] Examples of electron-donative coloring agents that can be used in the present invention include triphenyl-methane-phthalides, fluoranes, phenothiazines, indolylphthalides, spiropyranes, leuco-oramines [?] [translit.—Tr.] and rhodaminelactams, etc. Examples of such compounds include crystal violet lactone, rhodaminelactone, 3-diethylamino-6,8-dimethylfluorane, 1,3,3-trimethylindolino-2,2, spiro-6'-nitro-8'-methoxybenzopyrane, N-acetyloramine [translit.—Tr.] and benzoylleucomethylene blue, etc. These agents may be used singly or in combinations consisting of two or more agents. Various universally known compounds may be used as the abovementioned electron-donative coloring agents, as long as these compounds remain in the agent composition without being volatilized when the agent composition is used (i. e., as long as

these compounds are non-volatilizable compounds or compounds that are volatilized only with difficulty).

[0021] Such electron-donative coloring agents are generally difficult to dissolve in other chemical agents; if necessary, therefore, an auxiliary solvent may be used. However, in cases where the auxiliary solvent used as a desensitizing action, a compound with a boiling point that is lower than that of the volatilizable desensitizing agent that is employed should be used in order to prevent any effect on the color change of the coloring agent. Examples of such auxiliary solvents include ketones such as acetone and methyl ethyl ketone, etc., esters such as ethyl acetate, ethyl myristate and dimethyl phthalate, etc., ethers such as diethylene glycol dimethyl ether and triethylene glycol dimethyl ether, etc., alcohols such as n-octanol, etc., amines such as triethylamine, etc., and hydrocarbons such as toluene and xylene, etc.

[0022] Furthermore, there are no particular restrictions on the volatilizable desensitizing agent used in the present invention, as long as this agent has a desensitizing effect on the coloring of the abovementioned electron-donative coloring agent caused by the abovementioned inorganic developing compound, and is volatilized over time at ordinary temperatures. Examples of such volatilizable desensitizing agents include higher fatty acid esters such as isopropyl myristate, ethyl myristate and ethyl stearate, etc., and pyrethroids that are volatilizable at ordinary temperatures such as empentrin [translit.—Tr.], etc. One or more such agents may be used.

[0023] Furthermore, other miticides, fungicides and anti-mildew agents may be added if necessary to the indicator for insecticides provided by the present invention.

[0024] Furthermore, in the invention of Claim 2, the volatilizable desensitizing agent and electron-donative coloring agent are held in the inorganic developing compound itself or in a holding body which contains the inorganic developing compound, e. g., a solid agent configuration such as a mat, sheet or granular preparation prepared using an appropriate support or excipient. Accordingly, an indicator for insecticides which is simple and superior in terms of usability can be provided. Examples of such mats, sheets or granular supports include mats made of pulp, papers, woven fabrics, nonwoven fabrics, plastic molded articles made of polyethylene, polypropylene, polyvinyl chloride, polyester or ethylene – vinyl acetate, etc., and porous glass materials, etc. Such supports or excipients can be caused to contain the inorganic developing compound by methods such as coating, impregnation or kneading, etc., using various types of binders if necessary.

[0025] Furthermore, in the invention of Claim 3, a pyrethroid that is volatilizable at ordinary temperatures is used as the volatilizable desensitizing agent; as a result, this pyrethroid acts as both an active component and a volatilizable desensitizing agent. Accordingly, the composition is efficient, and there is a good correlation between the volatilization process of the active component and the color change. Examples of such pyrethroids that are volatilizable at ordinary temperatures include empentrin [translit.—Tr.], 5-propargyl-2-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate, 5-propargyl-2-methyl-3-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate, benfluthrin [translit.—Tr.], fenfluthrin [translit.—Tr.] and flametrin [sic] [translit.—Tr.], etc. Since one or two of these compounds are used, the volatilization process and endpoint of the volatilization process of the active component can be

detected very clearly, and an indicator for insecticides which has a high insecticidal/insect-proofing effect can be obtained.

[0026] Furthermore, in the invention of Claim 4, one or two compounds selected from a set consisting of empentrin [translit.—Tr.], 5-propargyl-2-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate, 5-propargyl-2-methyl-3-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate and benfluthrin [translit.—Tr.] are used as the abovementioned pyrethroid that is volatilizable at ordinary temperatures. Accordingly, an indicator for insecticides which is highly volatilizable at ordinary temperatures, and which combines a high insecticidal/insect-proofing effect, a low toxicity for warm-blooded animals and [good] chemical stability can be obtained.

[0027] In the indicator for insecticides provided by the present invention, the coloring of an electron-donative coloring agent caused by an inorganic developing compound is suppressed by the [abovementioned] volatilizable desensitizing agent, so that coloring is manifested as a result of the volatilization of the abovementioned agent. Accordingly, the coloring or lack of coloring of the electron-donative coloring agent is determined by the desensitizing effect of the abovementioned desensitizing agent and the strength of the developing action of the inorganic developing compound, so that the appropriate mixture proportions vary according to the combination of desensitizing agent and inorganic developing compound used. In the invention of Claim 5, the ratio of desensitizing agent to inorganic developing compound at which coloring occurs when the abovementioned volatilizable desensitizing agent is used as a desensitizing agent varies somewhat according to the type of inorganic developing compound used, but is generally in the range of 30/1 to 2/1. Accordingly, an indicator for insecticides is obtained in which the volatilization process and the endpoint of the volatilization process of the active component can be very clearly detected as a result of the coloring of the electron-donative coloring agent in this range.

[0028] Furthermore, in the invention of Claim 6, silica, activated alumina, clay, talc, powdered quartzite (powdered silica) [?] [doubtful wording—Tr.], acid clay, activated clay, bentonite, kaolin, cerite [?] [translit.—Tr.], fluorigel [?] [translit.—Tr.], pearlite, aluminum silicate, magnesium silicate, titanium oxide and zinc oxide are cited as examples of inorganic developing compounds, and coloring of the electron-donative coloring agent accompanying the volatilization of the desensitizing agent is manifested by using one or more of these compounds.

[0029] Furthermore, in the invention of Claim 7, one or more compounds selected from a set consisting of silica, activated alumina and fluorigel [translit.—Tr.] are used in particular as the abovementioned inorganic developing compound; accordingly, the developing action with respect to the electron-donative coloring agent is suitable, and an indicator for insecticides which shows a good color change is obtained.

[0030] Furthermore, in the invention of Claim 8, a pyrethroid that is volatilizable at ordinary temperature is used as the abovementioned volatilizable desensitizing agent, and one or more compounds selected from a set consisting of silica, activated alumina, clay, talc, powdered quartzite (powdered silica) [?], acid clay, activated clay, bentonite, kaolin, cerite [?] [translit.—Tr.], fluorigel [?] [translit.—Tr.], pearlite, aluminum silicate, magnesium silicate, titanium oxide

and zinc oxide are added as inorganic developing compounds; as a result, an indicator for insecticides in which the volatilization process and endpoint of the volatilization process of the pyrethroid that is volatilizable at ordinary temperatures can be very clearly detected is obtained.

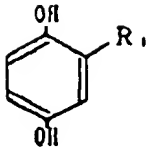
[0031] Furthermore, the invention of Claim 9 is characterized by the fact that in the invention of Claim 8, one or two compounds selected from a set consisting of empentrin [translit.—Tr.], 5-propargyl-2-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate, 5-propargyl-2-methyl-3-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate and benfluthrin [translit.—Tr.] are used as the abovementioned pyrethroid that is volatilizable at ordinary temperatures. Accordingly, an indicator for insecticides which combines a high insecticidal/insect-proofing effect, a low toxicity for warm-blooded animals and [good] chemical stability can be obtained.

[0032] Furthermore, the invention of Claim 10 is characterized by the fact that in the invention of Claim 8 or Claim 9, one or more compounds selected from a set consisting of silica, activated alumina and fluorigel [translit.—Tr.] are used in particular as the abovementioned inorganic developing compound; accordingly, the developing action with respect to the electron-donative coloring agent is suitable, and an indicator for insecticides which shows a good color change is obtained.

[0033] Furthermore, the invention of Claim 11 is characterized by the fact that in the invention of Claim 10 [Translator's note: in earlier descriptions of Claim 11, reference was also made to Claim 9, but only Claim 10 is mentioned here.] , [a] the abovementioned pyrethroid that is volatilizable at ordinary temperatures as the abovementioned volatilizable desensitizing agent consists of one or two compounds selected from a set consisting of empentrin [translit.—Tr.], 5-propargyl-2-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate, 5-propargyl-2-methyl-3-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate and benfluthrin [?] [translit.—Tr.], [b] the abovementioned inorganic developing compound consists of one or more compounds selected from a set consisting of silica, activated alumina and fluorigel [?], [c] the pyrethroid and developing compounds are mixed at a mixture ratio in the range of 100 : 1 ~ 5 : 1, and [d] [the indicator] shows developing properties together with the volatilization of the pyrethroid that is volatilizable at ordinary temperatures in the [mixture ratio] range of 30 : 1 ~ 2 : 1. Accordingly, an indicator for insecticides [a] in which coloring begins when the pyrethroid that is volatilizable at ordinary temperatures has been 50 to 80% volatilized, so that the volatilization process and endpoint of the volatilization process of the pyrethroid that is volatilizable at ordinary temperatures can be very clearly detected, and [b] which combines a high insecticidal/insect-proofing effect, a low toxicity for warm-blooded animals and [good] chemical stability, can be obtained.

[0034] Furthermore, the invention of Claim 12 is characterized by the fact that that in any of the inventions of Claims 8 through 11, the indicator contains a pyrethroid that is volatilizable at ordinary temperatures as the abovementioned volatilizable desensitizing agent, and said indicator also contains one or more hydroquinone type compounds expressed by general formula I

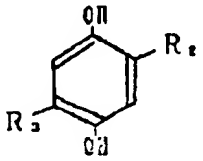
[Chemical Formula 5]



(in the above formula, R_1 indicates an alkyl group with 3 to 6 carbon atoms) as a stabilizer at the rate of 0.01 to 0.1 times the amount of the aforementioned pyrethroid that is volatilizable at ordinary temperatures. As a result, the stability of 5-propargyl-2-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate and 5-propargyl-2-methyl-3-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate in particular (among the abovementioned pyrethroids that are volatilizable at ordinary temperatures) is improved, so that an indicator for insecticides that is superior in terms of chemical stability and duration of potency is obtained. Examples of such stabilizers include mono-tertiary-butylhydroquinone and mono-tertiary-amylhydroquinone, etc.

[0035] Furthermore, the invention of Claim 13 is characterized by the fact that in the invention of Claim 12, one or more hydroquinone type compounds expressed by general formula II

[Chemical Formula 6]



(in the above formula, R_2 and R_3 indicate alkyl groups with 3 to 6 carbon atoms, which may be the same or different) are additionally added as stabilizers at the rate of 0.005 to 0.05 times the amount of the aforementioned pyrethroid that is volatilizable at ordinary temperatures. As a result, an indicator for insecticides that is even more superior in terms of chemical stability and duration of potency is obtained. Examples of such stabilizers include 2,5-di-tertiary-butylhydroquinone, etc.

[0036] Furthermore, in the invention of Claim 14, a volatilization-adjusting agent is used if necessary to adjust the volatilization rate of the aforementioned pyrethroid volatilizable at ordinary temperatures that is used in the present invention. Accordingly, the color change can be manifested more clearly. In this case, the volatilization-adjusting agent that is used may or may not be a desensitizing agent; however, in cases where this volatilization-adjusting agent does have a desensitizing action, it is necessary to take the abovementioned desensitizing agent/inorganic developing compound ratio into account. Furthermore, since the aforementioned stabilizers used for the pyrethroid are compounds that have phenolic hydroxy groups, the developing properties may be strengthened in some cases as a result of the addition of these compounds. In such cases, the color change can be appropriately adjusted by using agents that have a desensitizing effect. Examples of such volatilization-adjusting agents include diisobutyl phthalate, di-n-butyl adipate, diethyl sebacate, benzyl benzoate, isoamyl laurate, diethylene glycol di-n-butyl ether and triethylene glycol dimethyl ether, etc. These compounds may be used singly, or may be used in combinations consisting of two or more compounds. Furthermore, certain of these compounds may also be used as auxiliary solvents.

[0037] Furthermore, in the invention of Claim 15, a color change from one color to another color can be effected by adding a colored dye and/or a colored pigment beforehand along with the electron-donative coloring agent. Examples of such colored dyes and colored pigments include fast yellow G, benzidine yellow, indo-fast orange, irgadine [?] [translit.—Tr.] red, lake red C, rhodamine FB, phthalocyanine blue, brilliant green B, oil yellow GC, cayasette [?] [translit.—Tr.] YG, savon [?] [translit.—Tr.] fast orange RR and eisenspiron [?] [translit.—Tr.] red BEH, etc. One or more of these dyes or pigments may be used.

[0038] A method such as drip coating, immersion coating, spray coating, brush coating or printing, etc., on the holding body, or pasting to the holding body, may be utilized in the manufacture of the indicator of the present invention. Furthermore, in cases where the agents used are not liquid-form agents, or in cases where a solvent is not used, a mixture obtained by a method such as kneading or mixing and melting, etc., may be applied to the holding body by kneading, coating or printing, etc. Alternatively, the indicator may be formulated "as is" in gel form, powder form or granular form, etc. Furthermore, in cases where the mixture is applied to the holding body by coating, impregnation or printing, etc., this coating, impregnation or printing may be applied to the holding body overall, or may be applied in spot form, in a one-sided application or in the form of a pattern.

[0039] The concentration of the agent composition with respect to the holding body may be any concentration that causes the desired color change to occur, and that allows visual discrimination of this color change. This concentration depends on the type of holding body used, and on the color of the holding body itself; ordinarily, however, a concentration that is 10 to 80% of the saturation impregnation amount (or saturation content) of the holding body is suitable. If the amount of impregnation is too low, the diffusion of the agent composition into the holding body is poor, so that the color density of the color change is also low, thus making it difficult to judge the conditions of coloring. Conversely, if the holding body is impregnated with an amount that is close to the saturation impregnation amount, exudation of the agent composition occurs, so that there are problems of contamination, etc.

[0040] The indicator for insecticides provided by the present invention, which is obtained as described above, allows very clear visual detection of the volatilization process and endpoint of the volatilization process of the active component, and is highly stable.

[0041] Next, working examples and the results of tests of the effect of the present invention will be described in order to show the superiority of the present invention even more clearly.

[0042]

[Working Example 1] A mat with a thickness of 1.5 mm, a length of 20 mm and a width of 35 mm [sic] which had been surface-treated with a silica gel (used as an inorganic developing compound) at the rate of 4 mg per mat was impregnated with 40 mg of empentrin, 9.5 mg of diethylene glycol dimethyl ether and 0.5 mg of crystal violet lactone, thus producing an indicator for insecticides.

[0043]

[Working Example 2] A mat with a thickness of 1.5 mm, a length of 20 mm and a width of 35 mm which had been surface-treated with a silica gel (used as an inorganic developing compound) at the rate of 4 mg per mat was impregnated with 30 mg of empentrin, 50 mg of 5-propargyl-2-furylmethyl 2,2,3,3-tretramethylcyclopropanecarboxylate, 10 mg of triethylene glycol dimethyl ether, 5 mg of mono-tertiary-butylhydroquinone, 1 mg of 2,5-di-tertiary-butylhydroquinone and 1 mg of crystal violet lactone, thus producing an indicator for insecticides.

[0044]

[Working Example 3] A thick paper sheet with a thickness of 1 mm, a length of 60 mm and a width of 8 mm whose surface had been coated with activated alumina (used as an inorganic developing compound) at the rate of 20 mg per sheet was impregnated with 70 mg of empentrin, 160 mg of 5-propargyl-2-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate, 30 mg of diethylene glycol dimethyl ether and 3 mg of crystal violet lactone, thus producing an indicator for insecticides.

[0045]

[Test Example 1] A solution consisting of 40 parts by weight of the test volatilizable desensitizing agent, 1.0 parts by weight of crystal violet lactone and 59 parts by weight of acetone was partially poured [?] [uncertain wording—Tr.] into 50 mg of the test inorganic developing compound so that the ratio of the desensitizing agent to the developing compound was a specified ratio. The acetone was then dried by means of an air draft, and the color change was observed. The results obtained are shown in Table 1.

[0046]

[Table 1]

Desensitizing agent	Desensitizing agent/developing compound	Inorganic developing compound			
		Silica	Activated alumina	Activated clay	Diatomaceous earth
Empentrin	20/1	Δ	×-Δ	◎	×
	10/1	O	×-Δ	◎	×
	6/1	◎	O	◎	×
	4/1	◎	◎	◎	×
Dimethyl phthalate	20/1	×	×	-	-
	10/1	×-Δ	×	-	-
	6/1	×-Δ	×	-	-
	4/1	×-Δ	×-Δ	-	-

◎: Deeply colored
O: Colored
Δ: Slightly colored
×: No coloring seen

[0047] As a result of testing, it was found that when empentrin (a pyrethroid that is volatilizable at ordinary temperatures) was used as a volatilizable desensitizing agent, coloring occurred when the desensitizing agent/developing compound ratio was in the range of 20/1 ~ 4/1 in the case of silica and activated alumina, thus suggesting utility in terms of function as an indicator. In the case of dimethyl phthalate, on the other hand, almost no coloring occurred in the abovementioned range. Furthermore, when diatomaceous earth was used as the inorganic developing compound, the developing properties were too weak, so that no coloring was observed.

[0048]

[Test Example 2] A clothing insect-proofing mat (2 cm × 3.5 cm) prepared as in Working Example 1 was suspended inside a chest-of-drawers with a capacity of 600 liters, and the relationship between the change in the color of the mat that accompanied the volatilization of the active component at room temperature and amount of agent remaining was investigated. The results obtained are shown in Tables 2 and 3.

[0049]

[Table 2]

(Table 2)

		Composition (mg/mat)			Inorganic developing compound (mg/mat)	Amount of volatilizable desensitizing agent remaining (%)/color change		
		Pyrethroid volatilizable at ordinary temperatures	Other	Crystal violet lactone		1 month	4 months	6 months
Indicators of the present invention	1	Empentrin 40	Diethylene glycol dimethyl ether 9.5	0.5	Silica (2.0)	90/ Colorless	60/ Light blue	40/ Blue
	2	Empentrin 12 Teflametrin [translit.— Tr.] 28	" 9.5	0.5	" (")	90/ Light yellow	65/ Light blue-green	45/ Blue-green
	3	Empentrin 40	" 9.5	0.5	" (1.0)	90/ Colorless	60/ Very light blue	40/ Light blue

	4	Empentrin 40	" 9.5	0.5	Fluorigel [translit.— Tr.] (1.0)	90/ Colorless	60/ Very light blue	45/ Light blue
	5	Empentrin 40	" 9.5	0.5	Activated alumina (1.0)	90/ Colorless	60/ Light blue	40/ Blue
	6	Empentrin 12 Teflametrin 28	" 9.5	0.5	" (")	90/ Light yellow	65/ Light blue	45/ Blue
	7	Teflametrin 40	" 9.5	0.5	Clay (")	90/ Light yellow	65/ Light blue-green	45/ Blue-green
	8	Empentrin 40	" 9.5	0.5	Kaolin (")	90/ Colorless	60/ Light blue	40/ Blue
	9	Empentrin 40	" 9.5	0.5	Bentonite (")	90/ Colorless	60/ Light blue	40/ Blue
	10	Empentrin 40	" 9.5	0.5	Silica (0.2)	90/ Colorless	60/ Colorless	40/ Very light blue
	11	Empentrin 12 Teflametrin 28	" 9.5	0.5	" (10.0)	90/ Colorless	65/ Blue-green	45/ Blue-green
	12	Empentrin 12 Compound A 28	" 9.5	0.5	" (2.0)	90/ Light yellow	65/ Light blue-green	45/ Blue-green
	13	Empentrin 12 Benfluthrin 28	" 9.5	0.5	" (")	90/ Colorless	60/ Light blue-green	40/ Blue-green

[Table 3]

(Continuation of Table 2)

		Composition (mg/mat)			Inorganic developing compound (mg/mat)	Amount of volatilizable desensitizing agent remaining (%)/color change		
		Pyrethroid volatilizable at ordinary temperatures	Other	Crystal violet lactone		1 month	4 months	6 months
Indicators of the present invention	14	Compound A 40	Diethylene glycol dimethyl ether 9.5	0.5	Silica (2.0)	90/ Light yellow	65/ Light blue-green	45/ Blue-green
	15	Benfluthrin 40	" 9.5	0.5	" (")	90/ Colorless	60/ Light blue	40/ Blue
	16	Empentrin 12 Teflametrin 28	" 9.5 t-BHQ 2.0	0.5	" (")	90/ Light yellow	65/ Blue-green	45/ Blue-green
	17	Empentrin 12 Teflametrin 28	TEG-DME 9.5 t-BHQ 2.0	0.5	" (")	90/ Light yellow	70/ Light blue-green	50/ Blue-green
Comparative indicators	18	Empentrin 40	Diethylene glycol dimethyl ether 9.5	0.5	Nonyl phenol (5.0)	90/ Colorless	60/ Light blue	40/ Blue → colorless
	19	Empentrin 12 Teflametrin 28	Diethylene glycol dimethyl ether 9.5	0.5	" (")	90/ Light yellow	65/ Light blue-green	45/ Blue-green → colorless

[0050] In the tables, teflametrin [translit.—Tr.] indicates 5-propargyl-2-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate, which is a pyrethroid that is volatilizable at ordinary temperatures. Compound A indicates 5-propargyl-2-methyl-3-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate. TEG-DME indicates triethylene glycol dimethyl ether. t-BHQ indicates mono-tertiary-butylhydroquinone.

[0051] In the test results, the indicators for insecticides provided by the present invention showed a good correlation between the volatilization process of the active component and the color change of the mat, so that the amount of agent that remained could be very clearly detected by visual inspection. Furthermore, when the mixture ratio of the desensitizing agent to the developing compound was 200/1, a period of more than six months was required for a color change to take place; conversely, when the ratio of the desensitizing agent to the developing compound was 4/1, a color change began within six months, so that it was necessary to adjust the volatilization. Furthermore, in the case of indicators using nonylphenol as a developing agent, the color gradually disappeared following the color change, so that the composition again became colorless. On the other hand, in cases where mono-tertiary-butylhydroquinone was added as a stabilizer, the overall amount of the developing compound was increased, and the color change was slightly accelerated; however, it was possible to adjust the color change appropriately by using triethylene glycol dimethyl ether instead of diethylene glycol dimethyl ether.

[0052]

[Effect of the Invention] The indicator for insecticides provided by the invention of Claim 1 makes it possible to detect the volatilization process and endpoint of the volatilization process of the volatilizable desensitizing agent in a very clear and stable manner by visual inspection using the color change of an electron-donative coloring agent caused by an inorganic developing compound.

[0053] Furthermore, as a result of the invention of Claim 2, an indicator for insecticides can be provided by using the inorganic developing compound "as is" as a holding body for an agent composition consisting of a volatilizable desensitizing agent and an electron-donative coloring agent, or (usually more preferably) preparing the composition in a solid configuration such as a mat, sheet or granular configuration, etc., using an appropriate support or excipient.

[0054] Furthermore, as a result of the invention of Claim 3, the active component and the aforementioned volatilizable desensitizing agent can be combined by using a pyrethroid that is volatilizable at ordinary temperatures as the aforementioned volatilizable desensitizing agent; accordingly, an indicator for insecticides which is efficient and which shows a good correlation between the volatilization process of the active component and the color change can be obtained.

[0055] Furthermore, as a result of the invention of Claim 4, one or two compounds selected from a set consisting of empentrin, 5-propargyl-2-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate, 5-propargyl-2-methyl-3-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate and benfluthrin are used as the abovementioned pyrethroid that is volatilizable at ordinary temperatures; accordingly, an indicator for insecticides which combines a high volatilizability at ordinary temperatures, a high insecticidal/insect-proofing effect, a low toxicity for warm-blooded animals and [good] chemical stability can be obtained.

[0056] Furthermore, as a result of the invention of Claim 5, developing properties are shown when the ratio of desensitizing agent to inorganic developing compound is in the range of 30 : 1 to 2 : 1; accordingly, an indicator for insecticides is obtained in which the volatilization process

and the endpoint of the volatilization process of the active component can be very clearly detected.

[0057] Furthermore, as a result of the invention of Claim 6, examples of compounds that can be used as the aforementioned inorganic developing compound include silica, activated alumina, clay, talc, powdered quartzite (powdered silica) [?] [doubtful wording—Tr.], acid clay, activated clay, bentonite, kaolin, cerite [?] [translit.—Tr.], fluorigel [?] [translit.—Tr.], pearlite, aluminum silicate, magnesium silicate, titanium oxide and zinc oxide, and coloring of the electron-donative coloring agent accompanying the volatilization of the desensitizing agent is manifested by using one or more of these compounds. Accordingly, an indicator for insecticides in which the volatilization process and endpoint of the volatilization process of the active component can be very clearly detected is obtained.

[0058] Furthermore, as a result of the invention of Claim 7, silica, activated alumina or fluorigel [translit.—Tr.] is used in particular as the abovementioned inorganic developing compound; accordingly, the developing action with respect to the electron-donative coloring agent is suitable, and an indicator for insecticides which shows a good color change is obtained.

[0059] Furthermore, as a result of the invention of Claim 8, a pyrethroid that is volatilizable at ordinary temperature is used as the abovementioned volatilizable desensitizing agent, and one or more compounds selected from a set consisting of silica, activated alumina, clay, talc, powdered quartzite (powdered silica) [?], acid clay, activated clay, bentonite, kaolin, cerite [?] [translit.—Tr.], fluorigel [?] [translit.—Tr.], pearlite, aluminum silicate, magnesium silicate, titanium oxide and zinc oxide are added as inorganic developing compounds; consequently, an indicator for insecticides in which the volatilization process and endpoint of the volatilization process of the pyrethroid that is volatilizable at ordinary temperatures can be very clearly detected is obtained.

[0060] Furthermore, as a result of the invention of Claim 9, one or two compounds selected from a set consisting of empentrin [translit.—Tr.], 5-propargyl-2-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate, 5-propargyl-2-methyl-3-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate and benfluthrin [translit.—Tr.] are used as the abovementioned pyrethroid that is volatilizable at ordinary temperatures in the invention of Claim 8; accordingly, an indicator for insecticides which combines a high insecticidal/insect-proofing effect, a low toxicity for warm-blooded animals and [good] chemical stability can be obtained.

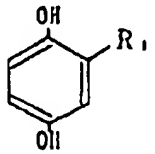
[0061] Furthermore, as a result of the invention of Claim 10, one or more compounds selected from a set consisting of silica, activated alumina and fluorigel [translit.—Tr.] are used in particular as the abovementioned inorganic developing compound in the invention of Claim 8 or Claim 9; accordingly, the developing action with respect to the electron-donative coloring agent is suitable, and an indicator for insecticides which shows a good color change is obtained.

[0062] Furthermore, as a result of the invention of Claim 11, [a] the abovementioned pyrethroid that is volatilizable at ordinary temperatures as the abovementioned volatilizable desensitizing agent consists of one or two compounds selected from a set consisting of empentrin [translit.—Tr.], 5-propargyl-2-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate, 5-propargyl-2-

methyl-3-furylmethyl 2,2,3,3-tetramethylcyclopropanecarboxylate and benfluthrin [?] [translit.—Tr.], [b] the abovementioned inorganic developing compound consists of one or more compounds selected from a set consisting of silica, activated alumina and fluorigel [?], [c] the pyrethroid and developing compounds are mixed at a mixture ratio in the range of 100 : 1 ~ 5 : 1, and [d] [the indicator] shows developing properties together with the volatilization of the pyrethroid that is volatilizable at ordinary temperatures in the [mixture ratio] range of 30 : 1 ~ 2 : 1, in the invention of [Claim 9 and] Claim 10; accordingly, an indicator for insecticides [a] in which coloring begins when the pyrethroid that is volatilizable at ordinary temperatures has been 50 to 80% volatilized, so that the volatilization process and endpoint of the volatilization process of the pyrethroid that is volatilizable at ordinary temperatures can be very clearly detected, and [b] which combines a high insecticidal/insect-proofing effect, a low toxicity for warm-blooded animals and [good] chemical stability, can be obtained.

[0063] Furthermore, as a result of the invention of Claim 12, the indicator contains a pyrethroid that is volatilizable at ordinary temperatures as the abovementioned volatilizable desensitizing agent, and said indicator also contains one or more hydroquinone type compounds expressed by general formula I

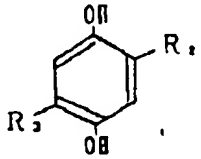
[Chemical Formula 7]



(in the above formula, R_1 indicates an alkyl group with 3 to 6 carbon atoms) as a stabilizer at the rate of 0.01 to 0.1 times the amount of the aforementioned pyrethroid that is volatilizable at ordinary temperatures, in the inventions of any of Claims 8 through 11; accordingly, an indicator for insecticides which is even more superior in terms of chemical stability and duration of potency, and which has a high insecticidal/insect-proofing effect, can be obtained.

[0064] Furthermore, as a result of the invention of Claim 13, one or more hydroquinone type compounds expressed by general formula II

[Chemical Formula 8]



(in the above formula, R_2 and R_3 indicate alkyl groups with 3 to 6 carbon atoms, which may be the same or different) are additionally added as stabilizers at the rate of 0.005 to 0.05 times the amount of the aforementioned pyrethroid that is volatilizable at ordinary temperatures in the invention of Claim 12; accordingly, an indicator for insecticides in which the chemical stability and duration of potency are improved even further can be obtained.

[0065] Furthermore, as a result of the invention of Claim 14, a volatilization-adjusting agent is used if necessary to adjust the volatilization rate of the aforementioned pyrethroid that is

volatilizable at ordinary temperatures, and especially in cases where a hydroquinone type compound that has phenolic hydroxy groups is used as a stabilizer for the aforementioned pyrethroid that is volatilizable at ordinary temperatures, a volatilization-adjusting agent that has a desensitizing effect is used; accordingly, an indicator for insecticides in which the color change is manifested more clearly can be obtained.

[0066] Furthermore, as a result of the invention of Claim 15, an indicator for insecticides in which there is a color change from one color to another color can be obtained by adding a colored dye and/or a colored pigment beforehand along with the aforementioned electron-donative coloring agent.